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(54) **Process for preparing thermoplastic elastomer laminates.**

(57) In accordance with the present invention, there are provided processes for preparing thermoplastic elastomer laminates, which comprise a step of obtaining a laminated sheet by placing a thermoplastic elastomer unexpanded sheet containing a polyolefin resin, an ethylene/α-olefin copolymer rubber and a foaming agent onto the back of a thermoplastic elastomer sheet containing a polyolefin resin and an ethylene/α-olefin copolymer rubber, and heat fusion bonding both sheets to each other, and a step of obtaining a thermoplastic elastomer laminate having a foamed layer by heating said laminated sheet to expand the thermoplastic elastomer unexpanded sheet layer thereof.

foamed PO + ethylene/α-olefin

PO + ethylene/α-olefin

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Description

PROCESS FOR PREPARING THERMOPLASTIC ELASTOMER LAMINATES

FIELD OF THE INVENTION

This invention relates to processes for preparing thermoplastic elastomer laminates and more particularly to processes for preparing soft thermoplastic elastomer laminates having a thin thermoplastic elastomer foamed layer, said laminates being useful for applications in automotive internal trim parts such as instrument panels and interior sheets, and in outer skin materials for bag.

BACKGROUND OF THE INVENTION

Thermoplastic elastomers have been widely used hitherto as outer skin materials for automotive internal trim and parts, for example, instrument panels and interior sheets. The thermoplastic elastomers possess such properties, both thermoplasticity and elasticity, and can be formed by injection molding, extrusion molding or the like into molded articles excellent in heat resistance, tensile properties, weatherability, flexibility and elasticity.

As concrete examples of the thermoplastic elastomers mentioned above, there are disclosed, for example, in Japanese Patent Publn. No. 34210/1978, thermoplastic elastomers obtained by dynamically partial curing a mixture comprising 60 - 80 parts by weight of monoolefin copolymer rubber and 40 - 20 parts by weight of polyolefin resin. Further, Japanese Patent Publn. No. 21021/1978 discloses thermoplastic elastomers comprising (a) partially crosslinked copolymer rubber containing ethylene/propylene/non-conjugated polyene copolymer rubber and having the gel content of 30 - 90% by weight, and (b) a polyolefin resin. Furthermore, Japanese Patent Publn. No. 18448/1980 discloses thermoplastic elastomers obtained by dynamically cross-linking ethylene/propylene copolymer and polyolefin resin, either partially or completely.

Sheets of the above-mentioned thermoplastic elastomers have heretofore been used as outer skin materials for automotive internal trim and parts such as instrument panels and interior sheets after bonding to or fusion integrating with core materials such as acrylonitrile/butadiene/styrene (ABS), polypropylene, aluminum and iron.

However, such sheets of the thermoplastic elastomers as mentioned above were found to be poor in soft touch as outer skins.

With view of solving the above-mentioned problem, such sheets of the thermoplastic elastomers used as outer skin materials were laminated on the back thereof with a polyethylene foam, polypropylene foam or urethane foam so that the cellular body will act as a pad, thereby imparting a soft touch to said sheets.

However, molded articles of foambacked thermoplastic elastomer sheets obtained by fabrication such as vacuum forming technique were lacking in sharpness of the shape and ridge thereof, though the polyethylene, polypropylene or urethane foam having an expansion coefficient of as large as 10 - 30 times was capable of imparting a soft touch to the thermoplastic elastomer sheets used as outer skin materials. In addition thereto, these foams as mentioned above were relatively expensive.

The present inventors found that when a thermoplastic elastomer sheet of a specific composition provided on the back thereof with a thin elastomer foam of a specific composition and a core material on which said elastomer sheet is laminated are formed by vacuum forming technique or the like to integrate them together, a molded article having a sharp shape, a sharp rudge and a soft touch is obtained, and they have eventually accomplished the present invention.

OBJECT OF THE INVENTION

The present invention is intended to solve such problems associated with the prior art as mentioned above, and an object of the invention is to provide processes for preparing thermoplastic elastomer laminates having a thin foam layer at a moderate cost by fabrication such as vacuum forming or compression molding technique, said thermoplastic elastomer laminates being excellent in sharpness of their shape and ridge and also excellent in soft touch.

SUMMARY OF THE INVENTION

The processes for preparing thermoplastic elastomer laminates of the present invention are characterized by comprising:

a step of obtaining a laminated sheet by placing on the back of a thermoplastic elastomer sheet comprising a polyolefin resin and an ethylene/α-olefin copolymer rubber a thermoplastic elastomer foamable sheet

comprising a polyolefin resin, an ethylene/α-olefin copolymer rubber and a foaming agent, followed by thermal fusion bonding, and

a step of obtaining a thermoplastic lastom r laminate having a foam layer by heating said laminated sheet, thereby foaming said thermoplastic elastomer foamable sheet.

DETAILED DESCRIPTION OF THE INVENTION

The processes for preparing thermoplastic elastomer laminates of the present invention are illustrated below in detail.

In the first step of the process of the present invention, a thermoplastic elastomer foamable sheet comprising a polyolefin resin, an ethylene/α-olefin copolymer rubber and a foaming agent is placed on the back of a thermoplastic elastomer sheet comprising a polyolefin resin and an ethylene/α-olefin copolymer rubber, and both sheets thus laid were thermal fusion bonded to each other to obtain a laminated sheet. In that case, it is desirable that the two sheets are thermal fusion bonded to each other so that the foamable sheet is not substantially foamed.

The thermoplastic elastomer constituting the thermoplastic elastomer sheet used in the present invention contains as essential components a polyolefin resin and an ethylene/α-olefin copolymer rubber.

In the present invention, useful as the above-mentioned ethylene/α-olefin copolymer rubber is a partially crosslinked product of ethylene/α-olefin copolymer rubber, however, a non-crosslinked product of ethylene/α-olefin copolymer rubber may also be used.

Preferred thermoplastic elastomers used in the invention may include, in concrete, such thermoplastic resin compositions as will be mentioned below.

(I) Thermoplastic resin compositions obtained by mixing various polyolefin resins represented by homopolymer of ethylene or propylene or copolymers thereof with small amounts of other polymeric monomers with partially crosslinked products of binary copolymer rubber comprising ethylene and α-olefin of 3 - 14 carbon atoms or ethylene/α-olefin copolymer rubber which is ternary or quaternary copolymer rubber obtained by copolymerizing said binary copolymer rubber with other various polyene compounds (see, for example, Japanese Patent Publn. No. 21021/1978 and Japanese Patent L-O-P Publn. No. 71738/1980).

(II) Thermoplastic resin compositions obtained by treating dynamically in the presence of small amounts of crosslinkers a blend of polyolefin resin and ethylene/α-olefin copolymer rubber (see, for example, Japanese Patent Publn. No. 34210/1978, Japanese Patent L-O-P Publns. No.149240/1978 and 149241/1978).

(III) Thermoplastic resin compositions obtained by blending the above-mentioned (I) or (II) further with polyolefin resin (see, for example, Japanese Patent L-O-P Publns. Nos. 145857/1978 and 1655/1979).

(IV) Thermoplastic resin compositions obtained heat treating dynamically in the presence of small amounts of peroxides a blend comprising peroxide crosslinking type polyolefin resin represented by homopolymer of ethylene or copolymer thereof with small amounts of other polymeric monomers, peroxide non-crosslinking type polyolefin resin represented by homopolymer of propylene or copolymer thereof with small amounts of other polymeric monomers, and ethylene/a-olefin copolymer rubber (see, for example, Japanese Patent L-O-P Publn. No. 71739/1980).

In these thermoplastic elastomers as illustrated above, the polyolefin resin and ethylene/α-olefin copolymer rubber are used in a weight ratio of usually 90/10 to 10/90, preferably 80/20 to 20/80.

As the polyolefin resin used in above cases, there may be used a mixture of polyethylene particularly low density polyethylene and polypropylene in a weight ratio of 10/90 to 70/30 for the purpose of improving moldability and scratch resistance.

From the standpoint of strength, it is desirable to use the ethylene/α-olefin copolymer rubber having an ethylene/α-olefin mole ratio of 50/50 to 90/10, preferably 70/30 to 85/15 and a Mooney viscosity ML_{1+4} (121°C) of at least about 20, preferably about 40 - 80. It is desirable that such ethylene/α-olefin copolymer rubber is partially crosslinked by dynamic heat treatment using generally about 0.1 - 2 parts by weight of an organic peroxide based on 100 parts by weight of the thermoplastic elastomer.

The thermoplastic elastomer used in the present invention may contain in the above-mentioned thermoplastic resin composition, if necessary, peroxide non-crosslinking type hydrocarbon rubbery substance represented by polyisobutylene, butyl rubber and the like and/or a mineral oil type softening agent.

In the present invention, the above-mentioned thermoplastic elastomer sheet is formed usually by T-die extrusion, calendering or the like forming technique.

The thermoplastic elastomer sheet used in the invention has a thickness of usually 0.05 - 5.0 mm, preferably 0.1 - 3.0 mm.

The thermoplastic elastomer constituting the thermoplastic elastomer foamable sheet used in the invention contains as an essential component a foaming ag nt in addition to said thermoplastic elastomer. The thermoplastic elastomer component constituting the thermoplastic elastomer foamable sheet, from which the foaming ag nt has been excluded, may be the same as or different from the thermoplastic elastomer component constituting the above-mentioned thermoplastic elastomer sheet.

Concrete examples of the foaming agent used in the invention include inorganic foaming agents such as

sodium hydrogencarbonate, sodium carbonat , ammonium hydrogen carbonate, ammonium carbonate, ammonium nitrite, etc.; nitroso compounds such as N,N′-dim thyl-N,N′-dinitrosoterephthalamide, N,N′-dinitrosopentamethylene tetramine, etc.; azo compounds such as azodicarbonamide, azobisisobutyronitrile, azocyclohexylnitrile, azodiaminobenzene, barium azodicarboxylate, etc.; sulfonyl hydrazide compounds such as benzensulfonyl hydrazide, toluenesulfonyl hydrazide, p,p′-oxybis(benzenesulfonylhydrazide), diphenylsulfone-3,3′-disulfonylhydrazide, etc.; and azide compounds such as calcium azide, 4,4′-diphenyldisulfonyl azide, p-toluenesulfonyl azide, etc. Of these compounds as exemplified above, preferred is azodicarbonamide.

In the present invention, the proportion of the foaming agent contained in the thermoplastic elastomer foamable sheet is 0.1 -50% by weight, preferably 0.5 - 20% by weight.

The thermoplastic elastomer formable sheet used in the invention is usually formed by T-die extrusion, calendering or the like forming technique.

The above-mentioned thermoplastic elastomer foamable sheet has a thickness of usually 0.05 - 5.0 mm, preferably 0.1 - 3.0 mm.

In the present invention, a laminated sheet is obtained by thermal fusion bonding the thermoplastic elastomer sheet, on the back of the thermoplastic elastomer foamable sheet, under such conditions where said foamable sheet does not foam. Usually in obtaining the laminated sheet, the thermoplastic elastomer sheet and the thermoplastic elastomer foamable sheet are individually brought to a molten state by heating at about 150 - 250°C using infrared radiation, and then both sheets are contact bonded to each other by using a hot roll kept at a temperature of about 30 - 100°C.

Subsequently, in the second step of the process of the present invention, the above-mentioned laminated sheet is heated to expand the thermoplastic elastomer foamable sheet layer, thereby obtaining a thermoplastic elastomer laminate having a foamed layer. In this case, the laminated sheet is preferably heated in an air oven, but other heating means may also be adaptable therefor.

The conditions under which the above-mentioned laminated sheet is heated, though said conditions may vary according to the kind of foaming agent used, are such that the temperature employed is usually 160 - 260°C, preferably 170 - 250°C and the heating time is 0.5 - 10 minutes, preferably 2 - 5 minutes.

In the present invention, an expansion coefficient of the foamable layer mentioned above is usually 1.1 - 20 times, preferably 1.1 - 8 times and particularly 1.2 - 5 times, and a thickness of the foamable layer is usually 0.1 - 10 mm, preferably 0.2 - 5 mm.

The thermoplastic elastomer laminate obtained in the present invention and a core material are integrated with each other by fabrication such as vacuum forming, hot press or compression molding technique, whereby a molded article which is sharp in shape and ridge and excellent in soft touch is obtained.

The core material to be laminated, if necessary, to the thermoplastic elastomer laminate prepared in the present invention includes a metallic plate, resin felt, paper, cloth, wood, glass or concrete in addition to the following thermoplastic resin.

The above-mentioned thermoplastic resin includes such resins, regardless of crystallinity or non-crystallinity, for example, polyolefins or ethylene/acrylic acid copolymers such as low density polyethylene, high density polyethylene, polypropylene, poly-1-butene, poly-4-methyl-1-pentene, or random or block copolymers of a-olefins such as ethylene, propylene, 1-butene and 4-methyl-1-pentene; ethylene/vinyl compound copolymers such as ethylene/vinyl acetate copolymers, ethylene/vinyl alcohol copolymers, ethylene/vinyl chloride copolymers, etc.; styrene resins such as polystyrene, acrylonitrole/styrene copolymers, ABS, methyl methacrylate/styrene copolymers, a-methylstyrene/styrene copolymers, etc.; polyacrylic acid esters such as polyvinyl chloride, polyvinylidene chloride, vinyl chloride/vinylidene chloride copolymers, methyl polymethacrylate, etc.; polyamides such as nylon 6, nylon 6/6, nylon 6/10, nylon 11, nylon 12, etc.; thermoplastic polyesters such as polyethylene terephthalate, polybutylene terephthalate, etc.; polycarbonates and polyphenylene oxides or mixtures thereof; or silicone or urethane type resins.

EFFECT OF THE INVENTION

The processes of the present invention have such effects that a relatively inexpensive thermoplastic elastomer laminate having a thin foamed layer is obtained, from which a molded article sharp in shape and ridge and excellent in soft touch can be obtained when said thermoplastic elastomer laminate is fabricated by such means as vacuum forming or compression molding technique.

The thermoplastic elastomer laminate is integrated, if necessary, with a core material, and the resulting laminate can be used for various applications, for example, automotive internal trim and parts such as instrument panel, interior sheets, etc. and outer skin materials for bag.

The present invention is illustrated below with reference to examples, but it should be construed that the invention is not limited to those examples.

Example 1

First, pellets of two kinds of thermoplastic elastomers, hereinafter called "TPE (A)" and "TPE (B)" respectively, were prepared in the following manner.

Preparation of pellets of TPE (A)

A mixtur comprising (i) 45 parts by w ight of isotactic polypropylene resin [melt flow rate: 13 g/10 min (230°C)], (ii) 55 parts by weight of ethylene/propylene/ethylidene norbornen terpolymer [ethylene unit/propylene unit (mole ratio): 78/22, iodine value: 15, Mooney viscosity ML_{1+4} (121°C) : 61], and (iii) 30 parts by weight of naphthene process oil was kneaded at 180°C for 5 minutes in a nitrogen atmosphere using a Banbury mixer, and the kneadate obtained was then formed with a sheet cutter into square pellets.

Subsequently, 100 parts by weight of the thus obtained square pellets and (iv) 1 part by weight of a mixture comprising 20% by weight of 1,3-bis(tert-butylperoxyisopropyl)benzene, 30% by weight of divinylbenzene and 50% by weight of paraffin mineral oil were kneaded together using a Henschel mixer and then extruded at 220°C in a nitrogen atmosphere using an extruder to prepare pellets of the thermoplastic elastomer TPE (A).

Preparation of pellets of TPE (B)

Pellets of the thermoplastic elastomer "TPE (B)" were prepared by repeating the same procedure as in the preparation of pellets of TPE (A) mentioned above except that (i) the amount of the isotactic polypropylene used was changed to 25 parts by weight, (ii) the amount of the ethylene/propylene/ethylidene norbornene terpolymer used was changed to 75 parts by weight, and further (v) 25 parts by weight of butyl rubber (a product of Esso, IIR 065) was added.

Separately, a sheet as an outer skin material for a laminated sheet was prepared from the above-mentioned TPE (A) and an unexpanded sheet for the laminated sheet was prepared from TPE (B), respectively in the following manner.

Preparation of TPE (A) sheet

The pellets of TPE (A) prepared above were extruded into a sheet-like form of 0.3 mm in thickness by using 90 mmø T-die extrusion molding machine manufactured and sold by Toshiba under such conditions that the screw used was full flight, L/D was 22, the extrusion temperature employed was 220°C, T-die used was a coathanger die and the take-off speed employed was 5 m/min, and the sheet-like form thus obtained was cooled by means of a cooling roll (roll temperature 35°C) to prepare the title sheet.

Preparation of TPE (B) unexpanded sheet

A mixture comprising 100 parts by weight of the pellets of TPE (B) prepared above, 2.0 parts by weight of azodicarbonamide (ADCA) as a forming agent, and 0.1 part by weight of triallylisocyanurate (TAIC) was kneaded by means of a Henschel mixer, and then from the resulting mixture, TPE (B) unexpanded sheet was prepared according to the procedure employed in the preparation of the above-mentioned TPE (A) sheet, provided that the extrusion temperature employed was 180°C and the thickness of the sheet was 0.5 mm.

Subsequently, the above-mentioned TPE (A) sheet was laminated to the above-mentioned TPE (B) unexpanded sheet in the following manner by using a laminator to prepare a laminated sheet.

Preparation of laminated sheet

The above-mentioned TPE (A) sheet and TPE (B) unexpanded sheet were individually preheated to 180°C by means of a preheater provided in the laminator, and thereafter both sheets are heat bonded to each other at a take-off speed of 5 m/min by means of two hot rolls and one cooling roll to obtain a laminated sheet.

Finally, the laminated sheet thus obtained was heated in the following manner to obtain a thermoplastic elastomer laminate having a foamed layer.

Preparation of thermoplastic elastomer laminate

The above-mentioned laminated sheet was allowed to stand in an air oven at 240°C for 4 minutes to expand the TPE (B) unexpanded sheet layer of said laminated sheet, whereby a thermoplastic elastomer laminate of 1.3 mm in thickness was obtained.

The outer skin layer and foamed layer constituting the thus obtained thermoplastic elastomer laminate had a thickn ss of 0.3 mm and of 1.0 mm, respectively. The thermoplastic elastomer laminate was evaluated with respect to tensile characteristics, surface hardness, torsion modulus, permanent elongation, heat aging characteristics and soft touch in the following manner. The evaluation sample was collected from the above-mentioned laminate by die-punching.

[Evaluation method]

Tensile characteristics	: Evaluated in accordance with JIS K 6301.
Surface hardness	: Evaluated in accordance with JIS A type of JIS K 6301.
Torsion modulus	: Evaluated in accordance with ASTM 1043.
Permanent elongation	: Evaluated in accordance with JIS K 6301.
Heat aging characteristics	: Evaluated in accordance with Tensile Test Method stipulated in JIS K 6301, using the evaluation sample allowed to stand at 120°C for 1,000 hours.
Soft touch	: Evaluated according to the following five-point ratings. 5 ... Very soft 4 ... Soft 3 ... Ordinary 2 ... Hard 1 ... Very hard

Results obtained in the above evaluation are shown in Table 1.

Comparative Example 1

An unexpanded laminated sheet obtained in the same procedure as in Example 1 was evaluated according to the same evaluation method as in Example 1.

Results obtained in the above evaluation are shown in Table 1.

Example 2

A laminate was obtained by repeating the same procedure as in Example 1 except that the amount of azodicarbonamide used in the preparation of the TPE (B) unexpanded sheet was changed to 1.5 parts by weight. The laminate obtained was evaluated according to the same evaluation method as in Examle 1.

Results obtained in the above evaluation are shown in Table 1.

Example 3

A laminate was obtained by repeating the same procedure as in Example 1 except that the amount of azodicarbonamide used in the preparation of the TPE (B) unexpanded sheet was changed to 3.0 parts by weight. The laminate obtained was evaluated according to the same evaluation method as in Example 1.

Results obtained in the above evaluation are shown in Table 1.

Example 4

A laminate was obtained by repeating the same procedure as in Example 1 except that the TPE (A) sheet and TPE (B) unexpanded sheet were prepared by calender forming under the conditions as mentioned below. The laminate obtained was then evaluated according to the same evaluation method as in Example 1.

The conditions under which the TPE (A) sheet and TPE (B) unexpanded sheet were formed respectively were the same. That is, both sheets were prepared by using a calendering machine manufactured and sold by Nippon Roll K.K. at a resin temperature of 180°C and a take-off speed of 20 m/min.

Results obtained in the above evaluation are shown in Table 1.

Example 5

A laminate was obtained by repeating the same procedure as in Example 4 except that the amount of the foaming agent azodicarbonamide used was changed to 3.0 parts by weight. The laminate obtained was evaluated according to the same evaluation method as in Example 1.

Results obtained in the above evaluation are shown in Table 1.

Examples 6 - 8

Laminates were obtained respectively by repeating the same procedure as in Example 1 exc pt that in place of 100 parts by weight of the TPE (B) used in the preparation of the TPE (B) unexpanded sh et, there were used TPE (B) and linear low density polyethylene (LLDPE) in the proportion (weight ratio) as mentioned below. The laminates obtained were evaluated according to the same evaluation method as in Example 1.

Example 6 : TPE (B)/LLDPE = 95/5
Example 7 : TPE (B)/LLDPE = 90/10
Example 8 : TPE (B)/LLDPE = 80/20

Results obtained in the above evaluation are shown in Table 1.

TABLE 1

	Example 1	Compar. Ex. 1	Example 2	Example 3	Example 4	Example 5	Example 6	Example 7	Example 8
TPE(B)/LLDPE [wt part]	100/0	100/0	100/0	100/0	100/0	100/0	95/5	90/10	80/20
Amount of foaming agent [wt part]	2.0	2.0	1.5	3.0	2.0	3.0	2.0	2.0	2.0
Forming Process of the sheet	T-die	T-die	T-die	T-die	Calender	Calender	T-die	T-die	T-die
Coefficient of expansion [time]	2.0	0	1.5	3.0	2.0	3.0	2.0	2.0	2.0
Tensile strength:									
M_{100} [kg/cm^2]	15	40	30	10	15	10	15	20	25
T_3 [kg/cm^2]	90	125	100	50	80	40	80	90	100
E_3 [%]	700	750	730	400	700	450	700	710	780
Surface hardness (JIS A type)	50	60	60	50	50	50	65	65	70
Torsion modulus [kg/cm^3]	10	20	15	10	20	10	25	30	35
Parmanent elongation [%]	10	20	20	25	20	25	30	30	30
Heat aging elongation [%]	650	700	700	250	600	200	680	700	750
Soft touch	5	1	3	5	4	5	4	4	4

Claims

1. A process for preparing a thermoplastic elastomer laminate, which comprises;preparing a laminated sheet by heat fusion bonding a thermoplastic elastomer foamable sheet containing a polyolefin resin, an ethylene/α-olefin copolymer rubber and a foaming agent to the back of a thermoplastic elastomer sheet containing a polyolefin resin and an ethylene/α-olefin copolymer rubber, and
heating said laminated sheet to expand the thermoplastic elastomer foamable sheet layer thereof and provide a foamed layer.

2. A process according to claim 1 wherein heat fusion bonding is effected while maintaining said foamable sheet in a substantially unexpanded state.

3. A process according to claim 1 or 2 wherein the expansion coefficient of the expanded thermoplastic elastomer is 1.1 to 20 times.

4. A process according to claim 1, 2 or 3 which comprises the further step of laminating the thermoplastic elastomer laminate to a core material to provide a molded article.